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(54) Title: WOOD PRESERVATIVE COMPOSITIONS

(57) Abstract: An insecticidal and termiticidal wood preservative composition contains as an active ingredient a compound of the formula: (I) wherein Y is a moiety selected from halogen, substituted aromatic ring and/or aliphatic residue, and n= 1 to 4.

WOOD PRESERVATIVE COMPOSITIONS

Field of the Invention

The present invention relates to the preservation of wood products. More specifically, the invention relates to the preservation of wood products from the attack of termites.

Background of the Invention

The preservation of timber against the attack of termites and other insect pests is a major undertaking because of the great damage inflicted by insects to stored timber and to wood and wood products, e.g., wood that is part of buildings or outdoor constructions.

Bromophenols have not been used in order to protect wood products from termites. One preferred illustrative compound of this class is Tetrabromobisphenol A (hereinafter referred to as "TBBA") which is a fire-retardant material, widely employed for protecting plastics against fire. JP 61-6769 (Publication No. 55-159915) discloses the coating of a single wood plate with TBBA as a protection against mould.

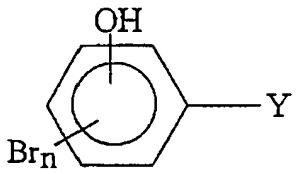
It is an object of the present invention to provide a family of brominated phenols that can be used for protecting wood and wood products against insect attack, particularly against termites.

It is another object of the invention to provide solutions of insecticidal and termiticidal bromophenols that can be used to impregnate wood or wood products.

Other purposes and advantages of the invention will appear as the description proceeds.

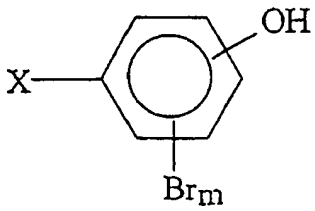
SUMMARY OF THE INVENTION

The invention is directed to an insecticidal and termiticidal wood preservative composition containing as an active ingredient a compound of the formula:

(I) 

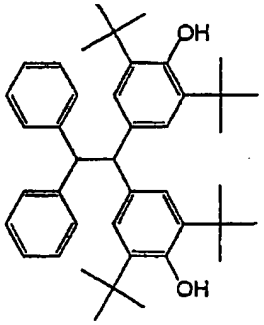
wherein Y is a moiety selected from halogen, substituted aromatic ring and/or aliphatic residue, and n= 1 to 4.

A preferred class of compounds useful in the composition of the invention in that in which Y is



wherein X is a moiety selected from alkylidene, NH, S or SO_2, and m= 0 to 4.

According to a preferred embodiment of the invention the compound of Formula (I) is selected from among tetrabromobisphenol A (TBBA), tribromobisphenol A (TBA), or a bromine derivative of a compound selected from Bisphenol F, Bisphenol A, Bisphenol S, Bisphenol Z, 4,4'-Biphenol, 1,1,2,2-tetra-(4-hydroxyphenyl) ethane, 2,2-hexafluoroisopropylidenediphenol, 1,1-ethylidenediphenol, 1,1-propylidenediphenol, or



The insecticidal and termiticidal composition of the invention is conveniently dissolved in a solvent prior to impregnation into the wood. Different solvents are suitable for this purpose. Illustrative and non-limitative preferred solvents are ethanol and aqueous basic solutions, e.g., in which the base is NaOH.

According to a preferred embodiment of the invention the active material is TBBA and its concentration ranges between 1% (w/w) and 20% (w/w), preferably between about 3% and 10%.

The composition of the invention may further comprise conventional termiticidal and insecticidal agents which are well known to the skilled person.

In another aspect the invention is directed to a method for protecting wood against termites, comprising impregnating the wood with a wood preservative composition as described above. The term "protection", as used herein, includes both the repulsion of termites away from the wood, and the actual killing thereof.

The present invention provides for a long-term protection of wood and wood products against termites by impregnating, e.g., by pressure-impregnation, the wood with an active ingredient, e.g. TBBA, the latter being dissolved either in an aqueous solvent or in an organic solvent or micro-emulsion. It should be appreciated that superficial coating of the wood with the active ingredient is not sufficient to achieve long-term protection, and therefore the invention is meant to relate to the in-depth impregnation of the wood with said active material. The actual depth of penetration of the active material will depend on many factors, as will

be apparent to the skilled person, such as the type of wood or wood product, the type of solution, the pressure employed, etc. However, the invention encompasses all cases in which at least some impregnation of the wood, below the surface, is achieved.

Detailed Description of the Invention

The aforementioned characteristics and advantages of the invention will be better understood through the following illustrative and non-limitative examples of preferred embodiments thereof.

Example 1

Method

The termiticidal and/or termite repellency effect of TBBA was examined by field exposure of TBBA treated timber samples to an active population of termites (***Macrotermes natalensis***). Much of the test detail (timber specimen species, dimensions, preservative impregnation method, scoring system etc.) was based on the European laboratory standard EN 117. This standard is used to define the toxic values of wood preservative formulations against wood destroying termites of the species ***Reticulitermes santonensis*** .

The TBBA active was formulated as a solvent-borne product for the test. (ethanol carrier). The precise preservative solutions used were 0%

(ethanol only), 1% TBBA, 3% TBBA, 5% TBBA, 7% TBBA and 10% TBBA (all w/w).

All wood block specimens were of Scots pine sapwood (*Pinus sylvestris*) with dimensions of 50 x 25 x 15 mm. All the test blocks were vacuum impregnated with TBBA according to the method of EN 117. Distribution of the treated blocks in the test was as follows:

Treated Specimens (TS):

3 (replicate specimens) x 5 (TBBA conc.s) = 15 (no.s 34-48)

Solvent Control Specimens (SCS):

3 (replicate specimens) x 1 (conc. (i.e. 0 % (ethanol only)) = 3 (no.s 49-51)

Untreated Control Specimens (UCS) (Virulence): = 3 (no.s 70-72)

TS Specimens: to determine the efficacy of the TBBA active (at different concentrations) against termite attack. SCS Specimens: to confirm absence of a termiticidal effect by the TBBA carrier (ethanol). UCS Specimens: to establish the virulence of the field population of termites.

The termite field site was established in Ginginglovu (Zululand, South Africa). The soil type of the area was a Hutton Bush Veldt soil (sandy loam soil). The site was situated within 10 m of an active termite mound on a raised flat platform of soil established some 5 cm above ground

level. The test specimens were randomly positioned on this platform and carefully covered with a waterproof flat plastic roof. This plastic roof was covered with soil to allow temperature and humidity conditions within the test zone to remain representative of the locale. The test specimens were left in-situ for a period of 54 days then scored for termite attack according the system defined in European standard EN 117 as follows:

- Score 0: No attack
- Score 1: Attempted attack
- Score 2: Slight attack
- Score 3: Average attack
- Score 4: Strong attack

Results

Table I displays mean termite attack scores (and high scores) for TBBA treated test specimens (ethanol carrier) after 54 days exposure to a field population of the termite *Macrotermes natalensis*. Table II displays mean termite attack scores (and high scores) for untreated virulence control specimens after 54 days exposure to a field population of the termite *Macrotermes natalensis*. In both tables, standard deviations are presented in the first parenthesis and the second parenthesis shows the highest score out of 3 replicates.

Table II shows a mean score of 2.67 (high score of 3) clearly indicating that the termite population at the site was very active and virulent. Table I shows that after 54 days exposure, of wood block specimens treated with solvent-borne TBBA at concentrations of 1%, 3%, 5%, 7% and 10%, only 1 block (of 3) treated with 7% had received any form of attack. The results for blocks treated with solvent only were severely attacked, indicating that the preservative carrier provided no protection. Under the scoring conditions of EN 117 therefore, the results indicate that the toxic values of solvent borne TBBA against termite attack lie between 1% and 10%.

Table I

TBBA Concentrations studied (%)	Treated Specimen Numbers	Mean Termite Attack Score (0-4)
10	34-36	0.00 (0.00) (0)
7	37-39	0.33 (0.58) (1)
5	40-42	0.00 (0.00) (0)
3	43-45	0.00 (0.00) (0)
1	46-48	0.00 (0.00) (0)
0	49-51	2.33 (0.58) (3)

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Table II

Treated Specimen Numbers	Mean Termite Attack Score (0-4)
70-72	2.67 (0.58) (3)

Example 2

Method

A laboratory test was undertaken to determine the toxic values of novel actives against termites. The active materials used were TBBA, TBBE [4,4'-ethylidenebis(2,6-dibromophenol)] [CAS RN = 12639-25-3] (Sample Number 1392-82-03); TBBZ [4,4'-cyclohexylidene(2,6-dibromophenol)] [CAS RN = 53350-96-2] (Sample Number 1392-72-03) and TBA, 2,2',6-tribromo-4,4'-isopropylidenediphenol, CAS 6386-73-8 (Sample Number 1392-75-03).

The test was undertaken largely according to the strictures laid down in ASTM 3345-74 (the American standard laboratory test to assess the termite resistant properties of wood to wood destroying termites). The test

method allows for the exposure of treated buried wood blocks to termites (*Reticulitermes* or *Coptotermes* spp.) over a period of 4 weeks. Efficacy of the product is determined by the extent of termite damage to the treated timber specimens through a scoring system. The test was slightly modified from the standard ASTM test by reducing the number of replicate wood specimens from 5 to 4. In addition, the ASTM scoring system was replaced by the more objective scoring system of the similar European standard EN 117 as follows:

Score 0: No attack

Score 1: Attempted attack

Score 2: Slight attack

Score 3: Average attack

Score 4: Strong attack

The toxic values of the preservative product (in kg/m^3 of wood) fall between two values as follows:

1) The lowest concentration which protects the wood (i.e. the concentration at which none of the test specimens show a degree of attack greater than 1).

2) The next lowest concentration in the series used at which the wood is no longer sufficiently protected (i.e. the concentration at which at least one test specimen shows a degree of attack of 2 or greater).

The validity of the test is confirmed if at least 2 of the untreated control specimens and at least 2 of the solvent/diluent control specimens correspond to a damage score of 4, and the corresponding colonies have at least 50% survivors.

The termites employed were *Coptotermes formosanus*.

The treatment solutions tested were:

- TBBA (Ethanol Carrier): 10%, 7%, 5%, 3% and 1%
- TBBE (Ethanol Carrier): 10%, 7%, 5%, 3% and 1%
- TBBZ (Ethanol Carrier): 10%, 7%, 5%, 3% and 1%
- TBA (Ethanol Carrier): 10%, 7%, 5%, 3% and 1%
- Ethanol Control

All wood bock specimens were of Southern Yellow Pine sapwood (*Pinus* spp.) with dimensions of 25.4 x 25.4 x 6.4 mm in accordance with ASTM D3345-74. All blocks were vacuum impregnated with the relevant

solutions in accordance with ASTM D3345-74. Distribution of blocks in the test was as follows:

TBBA:	4 (specimens) x 5 (conc.) = 20 (block no.s 21-40)
TBBE:	4 (specimens) x 5 (conc.) = 20 (block no.s 61-80)
TBBZ:	4 (specimens) x 5 (conc.) = 20 (block no.s 81-100)
TBA:	4 (specimens) x 5 (conc.) = 20 (block no.s 101-120)
Ethanol Control:	4 (specimens) = 4 (block no.s 181-184)

Ethanol controls were used to establish both the absence of any termicidal effect due to the ethanol carrier and the virulence of the termites used in the test.

The test specimens were conditioned and subjected to a leaching schedule (after vacuum impregnation) as stipulated in ASTM D1413-76 (as recommended in ASTM D3345-74). All test specimens were then air dried for two weeks to constant weight.

The specimens were each positioned at the base of a 500ml polyethylene culture jar (numbered according to each test specimen) and covered with 200g of sand (pre-washed, sterilised and air conditioned in the laboratory) to which was added 25g of de-ionised water. A further two containers containing watered sand but no timber were also prepared. All the jars

were sealed (lids pre-pierced and covered with stainless steel mesh for aeration).

Two days after specimen placement, 200 termites were added to each culture jar (90% workers and 10% soldiers) and the jars re-sealed. The jars were incubated at a mean temperature and relative humidity of 27.5°C and 75-90% for 4 weeks. The containers were provided with an artificial lighting regime of 10 hours light and 14 hours darkness.

After 1 week and 4 weeks each container was examined to determine termite mortality and normal tunneling behaviour patterns as follows:

- Is there tunneling present: YES/NO
- Are there any termites on the surface of the soil: YES/NO (if yes how many, i.e. 1-25% of pop, 26-50% of pop, 51-100% of pop.)
- Are any dead termites visible: YES/NO (if yes how many, 1-33%, 34-66%, 67-100%)

After 4 weeks the test specimens were removed and cleaned of debris. Each block was examined and visually rated for termite damage using the scoring system detailed above.

Results

Table III shows termite attack ratings on wood specimens treated with ethanol after 4 weeks incubation in sand culture.

Tables IV, V, VI and VII show termite attack ratings on wood specimens treated with TBBA, TBBE, TBBZ and TBA respectively, after 4 weeks incubation in sand culture.

Table IIIa shows tunneling activity, surface activity and mortality of termites after 1 and 4 weeks exposure in sand culture to wood specimens treated with ethanol.

Tables IVa, Va, VIa and VIIa show tunneling activity, surface activity and mortality of termites after 1 and 4 weeks exposure in sand culture to wood specimens treated with TBBA, TBBE, TBBZ and TBA respectively.

Ethanol Specimens: Table III shows that the termites used in the test were virulent (and were not affected by ethanol treatment of these specimens) with 2 of the 4 test specimens suffering strong attack. This was to be expected as table IIIa shows that the termites thrived under the test conditions with tunneling activity and complete survival noted throughout the test period.

TBBA Specimens: Table IV shows that the toxic values of TBBA are 32.52 and 44.00 kg/m^3 (solution concentrations of 5 and 7%). The score of 2 for one of the test specimens, at a mean loading of 55.26 kg/m^3, is disregarded as an anomalous result. The surface activity data in table IVa indicates that the two highest TBBA concentrations (7 and 10%) resulted in a degree of termite repellency throughout the incubation period. In addition, the three highest concentrations (5, 7 and 10%) resulted in very significant termite mortality (generally 67-100%) by the end of the incubation period.

TBBE Specimens: Table V shows that the toxic values of TBBE are 28.53 and 35.93 kg/m^3 (solution concentrations of 5 and 7%). The surface activity noted in table Va indicates that TBBE provided a repellent effect at certain stages of the incubation period. The highest concentration of TBBE (10%) resulted in 67-100% termite mortality by the end of the incubation period.

TBBZ Specimens: The presence of individual ratings of 2 at the three highest concentrations prevented the determination of toxic values for this active (Table VI). However, it is clear that the higher concentrations do reduce termite attack. TBBE also provided a repellent effect early in the incubation period at higher concentrations and resulted

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in significant termite mortality by the end of the incubation period (table VIa).

TBA Specimens: Table VII shows that the toxic values of TBA are 30.17 and 41.22 kg/m^3 (solution concentrations of 5 and 7%). TBA produced no repellent effect on the termite population but the higher concentrations resulted in 34-66% termite mortality by the end of the incubation period (table VIIa).

Table III

Specimen No.	Termite Attack Rating	Mean Rating
181	3	3.50 (0.58)
182	4	
183	4	
184	3	

Table IV

Specimen No.	Solution Conc. (%)	Loading (kg/m³)	Mean (kg/m³)	Termite Attack Rating	Mean Rating
21	10	48.0565		1	
22	10	56.3233	54.28	1	1.00
23	10	59.6759	(4.95)	1	(0.00)
24	10	53.0682		1	
25	7	39.6153		1	
26	7	35.6604	35.93	1	1.00
27	7	33.1234	(2.70)	1	(0.00)
28	7	35.3348		1	
29	5	30.0848		1	
30	5	28.1562	28.53	1	1.50
31	5	25.3130	(2.39)	2	(0.58)
32	5	30.5730		2	
33	3	17.7648		2	
34	3	16.4121	17.78	2	2.25
35	3	17.8172	(1.11)	3	(0.50)
36	3	19.1350		2	
37	1	7.5465		3	
38	1	6.8405	6.94	2	2.75
39	1	6.6395	(0.41)	4	(0.95)
40	1	6.7444		2	

Table V

Specimen No.	Solution Conc. (%)	Loading (kg/m³)	Mean (kg/m³)	Termite Attack Rating	Mean Rating
61	10	51.7924		1	
62	10	59.9261	55.26	1	1.25
63	10	49.5835	(5.35)	1	(0.50)
64	10	59.7234		2	
65	7	42.7084		1	
66	7	46.2477	44.00	1	1.00
67	7	42.4744	(1.77)	1	(0.00)
68	7	44.5619		1	
69	5	33.4407		2	
70	5	30.9586	32.52	1	1.50
71	5	33.1791	(1.11)	1	(0.58)
72	5	32.5056		2	
73	3	17.1295		2	
74	3	17.4279	17.94	2	2.00
75	3	17.6150	(1.12)	3	(0.82)
76	3	19.5934		1	
77	1	5.7355		3	
78	1	5.8468	5.62	3	3.00
79	1	5.5676	(0.23)	3	(0.00)
80	1	5.3174		3	

NB: 0: No Attack / 1: Attempted Attack / 2: Slight Attack / 3: Average Attack / 4: Strong Attack

Table VI

Specimen No.	Solution Conc. (%)	Loading (kg/m^3)	Mean (kg/m^3)	Termite Attack Rating	Mean Rating
81	10	56.1953		1	
82	10	56.5116	**56.09**	1	1.25
83	10	55.8560	(0.34)	1	(0.50)
84	10	55.7841		2	
85	7	37.8957		1	
86	7	35.8088	**39.36**	1	1.25
87	7	43.1990	(3.17)	2	(0.50)
88	7	40.1507		1	
89	5	29.1654		1	
90	5	30.1080	**28.54**	1	1.25
91	5	28.2496	(1.47)	1	(0.50)
92	5	26.6558		2	
93	3	18.8209		2	
94	3	18.9218	**16.96**	1	1.50
95	3	13.8822	(2.40)	2	(0.58)
96	3	16.2365		1	
97	1	5.6620		4	
98	1	5.0941	**5.95**	2	2.50
99	1	6.4104	(0.71)	2	(1.00)
100	1	6.6534		2	

NB: 0: No Attack / 1: Attempted Attack / 2: Slight Attack / 3: Average Attack / 4: Strong Attack

Table VII

Specimen No.	Solution Conc. (%)	Loading (kg/m³)	Mean (kg/m³)	Termite Attack Rating	Mean Rating
101	10	62.5923		1	
102	10	56.2025	55.10	1	1.00
103	10	54.1902	(6.25)	1	(0.00)
104	10	47.4157		1	
105	7	40.2913		1	
106	7	37.8605	41.22	1	1.00
107	7	44.5498	(2.84)	1	(0.00)
108	7	42.1750		1	
109	5	29.4314		2	
110	5	31.2399	30.17	1	1.75
111	5	30.1471	(0.78)	2	(0.50)
112	5	29.8682		2	
113	3	17.8751		2	
114	3	18.3913	18.41	2	1.75
115	3	20.7329	(1.71)	1	(0.50)
116	3	16.6446		2	
117	1	5.2534		3	
118	1	6.1841	5.82	2	3.00
119	1	5.3939	(0.58)	3	(0.82)
120	1	6.4328		4	

Table IIIa

Specimen No.	Tunneling Activity (1 week)	Surface Activity (1 week)	Visible Mortality (1 week)
181	Yes	No	No
182	Yes	No	No
183	Yes	No	No
184	Yes	No	No
Specimen No.	**Tunneling Activity (4 weeks)**	**Surface Activity (4 weeks)**	**Visible Mortality (4 weeks)**
181	Yes	No	No
182	Yes	No	No
183	Yes	No	No
184	Yes	No	No

Table IVa

Specimen No.	Tunneling Activity (1/4 weeks)	Surface Activity (1/4 weeks)	Visible Mortality (1/4 weeks)
21	Yes/Yes	No/1-25% (of pop.)	No/67-100% (of pop.)
22	Yes/Yes	No/1-25% (of pop.)	No/67-100% (of pop.)
23	Yes/Yes	No/1-25% (of pop.)	No/67-100% (of pop.)
24	Yes/Yes	No/1-25% (of pop.)	No/67-100% (of pop.)
25	Yes/Yes	No/No	No/1-33% (of pop.)
26	Yes/Yes	No/No	No/1-33% (of pop.)
27	Yes/Yes	No/No	No/1-33% (of pop.)
28	Yes/Yes	No/No	No/1-33% (of pop.)
29	Yes/Yes	1-25%/No (of pop.)	1-33%/1-33% (of pop.)
30	Yes/Yes	1-25%/No (of pop.)	1-33%/1-33% (of pop.)
31	Yes/Yes	1-25%/No (of pop.)	No/1-33% (of pop.)
32	Yes/Yes	1-25%/No (of pop.)	No/1-33% (of pop.)
33	Yes/Yes	No/No	No/1-33% (of pop.)
34	Yes/Yes	No/No	No/1-33% (of pop.)
35	Yes/Yes	No/No	No/No
36	Yes/Yes	No/No	No/No
37	Yes/Yes	No/No	No/No
38	Yes/Yes	No/No	No/No
39	Yes/Yes	No/No	No/No
40	Yes/Yes	No/No	No/No

Table Va

Specimen No.	Tunneling Activity (1/4 weeks)	Surface Activity (1/4 weeks)	Visible Mortality (1/4 weeks)
61	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
62	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
63	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
64	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
65	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
66	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
67	Yes/Yes	1-25%/1-25% (of pop.)	1-33%/67-100% (of pop.)
68	Yes/Yes	1-25%/1-25% (of pop.)	No/67-100% (of pop.)
69	Yes/Yes	No/No	No/67-100% (of pop.)
70	Yes/Yes	No/No	No/67-100% (of pop.)
71	Yes/Yes	No/No	No/34-66% (of pop.)
72	Yes/Yes	No/No	No/1-33% (of pop.)
73	Yes/Yes	No/No	No/1-33% (of pop.)
74	Yes/Yes	No/No	No/1-33% (of pop.)
75	Yes/Yes	No/No	No/1-33% (of pop.)
76	Yes/Yes	No/No	No/1-33% (of pop.)
77	Yes/Yes	No/No	No/No
78	Yes/Yes	No/No	No/No
79	Yes/Yes	No/No	No/No
80	Yes/Yes	No/No	No/No

Table VIa

Specimen No.	Tunneling Activity (1/4 weeks)	Surface Activity (1/4 weeks)	Visible Mortality (1/4 weeks)
81	Yes/Yes	1-25%/No (of pop.)	No/67-100% (of pop.)
82	Yes/Yes	1-25%/No (of pop.)	No/67-100% (of pop.)
83	Yes/Yes	1-25%/No (of pop.)	No/67-100% (of pop.)
84	Yes/Yes	1-25%/No (of pop.)	No/67-100% (of pop.)
85	Yes/Yes	1-25%/No (of pop.)	No/34-66% (of pop.)
86	Yes/Yes	1-25%/No (of pop.)	No/34-66% (of pop.)
87	Yes/Yes	1-25%/No (of pop.)	No/34-66% (of pop.)
88	Yes/Yes	1-25%/No (of pop.)	No/34-66% (of pop.)
89	Yes/Yes	No/No	No/34-66% (of pop.)
90	Yes/Yes	No/No	No/34-66% (of pop.)
91	Yes/Yes	No/No	No/34-66% (of pop.)
92	Yes/Yes	No/No	No/34-66% (of pop.)
93	Yes/Yes	No/No	No/1-33% (of pop.)
94	Yes/Yes	No/No	No/1-33% (of pop.)
95	Yes/Yes	No/No	No/1-33% (of pop.)
96	Yes/Yes	No/No	No/1-33% (of pop.)
97	Yes/Yes	No/No	No/No
98	Yes/Yes	No/No	No/No
99	Yes/Yes	No/No	No/No
100	Yes/Yes	No/No	No/No

Table VIIa

Specimen No.	Tunneling Activity (1/4 weeks)	Surface Activity (1/4 weeks)	Visible Mortality (1/4 weeks)
101	Yes/Yes	No/No	No/34-66% (of pop.)
102	Yes/Yes	No/No	No/34-66% (of pop.)
103	Yes/Yes	No/No	No/34-66% (of pop.)
104	Yes/Yes	No/No	No/34-66% (of pop.)
105	Yes/Yes	No/No	No/34-66% (of pop.)
106	Yes/Yes	No/No	No/34-66% (of pop.)
107	Yes/Yes	No/No	No/34-66% (of pop.)
108	Yes/Yes	No/No	No/34-66% (of pop.)
109	Yes/Yes	No/No	No/34-66% (of pop.)
110	Yes/Yes	No/No	No/34-66% (of pop.)
111	Yes/Yes	No/No	No/34-66% (of pop.)
112	Yes/Yes	No/No	No/34-66% (of pop.)
113	Yes/Yes	No/No	No/1-33% (of pop.)
114	Yes/Yes	No/No	No/1-33% (of pop.)
115	Yes/Yes	No/No	No/1-33% (of pop.)
116	Yes/Yes	No/No	No/1-33% (of pop.)
117	Yes/Yes	No/No	No/No
118	Yes/Yes	No/No	No/No
119	Yes/Yes	No/No	No/No
120	Yes/Yes	No/No	No/No

From the reported results, the following conclusions may, *inter alia*, be reached:

1) Impregnation of test specimens with concentrated solutions of TBBA, TBBE, TBBZ and TBA, followed by leaching, resulted in a significant reduction in termite attack.

2) Impregnation of test specimens with concentrated solutions of TBBA, TBBE and TBBZ resulted in a degree of termite repellency and the highest treatment concentrations of

TBBA, TBBE, TBBZ and TBA generally resulted in 67-100% mortality of the termite population by the end of the trial.

Example 3

Method

A further experiment was undertaken according to the modified ASTM 3345-74 test detailed in Example 2 with the following 6 changes to the protocol:

1. The solutions tested were as follows:
   - TBBA (NaOH Carrier): 10%, 7%, 5%, 3% and 1%
   - TBBF (NaOH Carrier): 10%, 7%, 5%, 3% and 1%
   - TBBS (NaOH Carrier): 10%, 7%, 5%, 3% and 1%
   - NaOH Carrier Control: 10%, 7%, 5%, 3% and 1%
   - H_2O Control
   - Untreated Control

2. The incubation substrate was a soil/sand mixture (1:1) to which had been added 75 ml of de-ionised water. This substrate was chosen as being more representative of the laboratory conditions under which the wild caught termites (***C. formosanus***) were maintained.
3. The treated timber samples were positioned on plastic mesh on top of the soil substrate. This method of placement is that used in

European Standard EN 117 and is more representative of the feeding conditions of the wild caught termites maintained in the laboratory.

4. 250 termites were used in each incubation vessel.

5. The wood block specimens were not subjected to a leaching process.

6. Evidence of termite tunneling activity and mortality was determined at completion of the incubation period only (4 weeks) and no examinations were undertaken for surface activity.

Distribution of blocks in the test was as follows:

TBBA: 4 (specimens) x 5 (conc.) = 20 (block no.s 41-60)

TBBF: 4 (specimens) x 5 (conc.) = 20 (block no.s 121-140)

TBBS: 4 (specimens) x 5 (conc.) = 20 (block no.s 141-160)

NaOH Carrier Control: 1 (specimen) x 5 (conc.) = 5 (blocks 163/165/172/174/179)

H_2O Control: 4 (specimens) = 4 (block no.s 185-188)

Untreated Control: 4 (specimens) = 4 (block no.s 189-192)

NaOH Carrier, H_2O and untreated controls were used to establish both the absence of any termiticidal effect due to the NaOH carrier solution and the virulence of the termites used in the test.

Results

Table VIII shows termite attack ratings on untreated wood specimens after 4 weeks incubation in soil/sand culture.

Table IX shows termite attack ratings on wood specimens treated with de-ionised water after 4 weeks incubation in soil/sand culture.

Tables X, XI and XII show termite attack ratings on wood specimens treated with TBBA, TBBF and TBBS respectively, after 4 weeks incubation in soil/sand culture.

Table XIII shows termite attack ratings on wood specimens treated with NaOH carrier solutions after 4 weeks incubation in soil/sand culture.

Table VIIIa shows tunneling activity and mortality of termites after 4 weeks exposure in soil/sand culture to untreated wood specimens.

Table IXa shows tunneling activity and mortality of termites after 4 weeks exposure in soil/sand culture to wood specimens treated with de-ionised water.

Tables Xa, XIa and XIIa show tunneling activity and mortality of termites after 4 weeks exposure in soil/sand culture to wood specimens treated with TBBA, TBBF and TBBS respectively.

Table XIIIa shows tunneling activity and mortality of termites after 4 weeks exposure in soil/sand culture to wood specimens treated with NaOH carrier solutions.

Control Specimens: Tables VIII and IX (untreated and water treated specimens) show that the termites used in the test were highly virulent with 3 of the 8 test specimens suffering the highest level of attack according to the rating system. In addition, Table XIII shows that none of the NaOH control specimens, except that treated with the highest concentration of NaOH, escaped the highest level of attack. Tables VIIIa, IXa and XIIIa show that the high level of attack experienced by all the foregoing test specimens was clearly due to the test conditions favoring termite survival with the great majority of the termites active at the end of the test period.

TBBA Specimens: Table X shows that the upper mean toxic value of TBBA, according to these test results, was 6.97 kg/m^3 (solution concentration of 1%). The data indicate that significant attack was prevented by all concentrations of TBBA used. Despite this low level of attack, table Xa shows that the majority of the termites were still active at the end of the test period. TBBA appeared therefore to be acting as a repellent throughout the trial period. This is supported by the observation that the termites had only partially soil covered those test specimens treated with 5, 7 and 10% solutions of TBBA, while those specimens treated with the lower concentrations were invariably completely covered.

TBBF Specimens: Table XI shows that the toxic values of TBBF are 6.78 and 20.53 kg/m^3 (solution concentrations of 1 and 3%). The data again indicate that significant attack was prevented by all concentrations of TBBF used. Table XIa shows that the majority of the termites were killed by the end of the test period indicating a significant termiticidal effect. However, as was found for TBBA, those test specimens treated with 5, 7 and 10% solutions of TBBF were only partially soil covered at the end of the test, thereby demonstrating a repellent effect also.

TBBS Specimens: Table XII shows that the upper mean toxic value of TBBA, according to these test results, was 7.27 kg/m^3 (solution concentration of 1%). As shown for both TBBA and TBBF, significant

attack was prevented by all concentrations of TBBS used. Table XIIa shows that exposure to TBBS resulted in significant termite mortality by the end of the test period. Once more a repellent effect was noted with test specimens treated with 5, 7 and 10% solutions of TBBS being partially soil covered at the end of the test.

Table VIII

Specimen No.	Termite Attack Rating	Mean Rating
189	4	
190	2	2.75
191	3	(0.96)
192	2	

Table IX

Specimen No.	Termite Attack Rating	Mean Rating
185	4	
186	2	3.00
187	4	(1.15)
188	2	

Table X

Specimen No.	Solution Conc. (%)	Loading (kg/m³)	Mean (kg/m³)	Termite Attack Rating	Mean Rating
41	10	74.6900		1	
42	10	65.7449	71.14	1	1.00
43	10	71.2935	(3.86)	1	(0.00)
44	10	72.8397		1	
45	7	49.5152		1	
46	7	54.1000	51.69	1	0.75
47	7	51.5702	(1.88)	0	(0.50)
48	7	51.5850		1	
49	5	35.8990		1	
50	5	36.4348	36.78	1	1.00
51	5	36.2667	(1.19)	1	(0.00)
52	5	38.5379		1	
53	3	20.9089		1	
54	3	23.6298	22.40	1	1.00
55	3	22.0586	(1.18)	1	(0.00)
56	3	22.9877		1	
57	1	7.1182		1	
58	1	7.1204	6.97	1	1.00
59	1	7.1712	(0.33)	1	(0.00)
60	1	6.4710		1	

NB: 0: No Attack / 1: Attempted Attack / 2: Slight Attack / 3: Average Attack / 4: Strong Attack

BNSDOCID: <WO_03009981A1_I_>

Table XI

Specimen No.	Solution Conc. (%)	Loading (kg/m³)	Mean (kg/m³)	Termite Attack Rating	Mean Rating
121	10	61.1175	56.07 (5.21)	1	0.5 (0.58)
122	10	54.6652		0	
123	10	49.3758		0	
124	10	59.1172		1	
125	7	45.6618	38.61 (5.62)	1	0.75 (0.50)
126	7	37.4983		0	
127	7	32.0221		1	
128	7	39.2380		1	
129	5	25.7958	29.91 (3.20)	0	0.5 (0.58)
130	5	33.5691		0	
131	5	30.5580		1	
132	5	29.7245		1	
133	3	21.1138	20.53 (1.61)	0	0.75 (0.50)
134	3	22.3233		1	
135	3	20.1881		1	
136	3	18.4960		1	
137	1	7.0290	6.78 (0.51)	3	1.50 (1.00)
138	1	6.6132		1	
139	1	6.1630		1	
140	1	7.3321		1	

NB: 0: No Attack / 1: Attempted Attack / 2: Slight Attack / 3: Average Attack / 4: Strong Attack

Table XII

Specimen No.	Solution Conc. (%)	Loading (kg/m³)	Mean (kg/m³)	Termite Attack Rating	Mean Rating
141	10	66.4361		0	
142	10	59.5467	68.25	0	0.25
143	10	75.9517	(6.98)	0	(0.50)
144	10	71.0662		1	
145	7	55.0388		1	
146	7	45.9981	48.81	1	0.75
147	7	39.3169	(7.61)	0	(0.50)
148	7	54.8711		1	
149	5	31.9904		1	
150	5	34.6792	34.70	1	1.00
151	5	35.5325	(1.96)	1	(0.00)
152	5	36.5564		1	
153	3	19.4698		1	
154	3	21.3925	20.11	0	0.5
155	3	21.5804	(1.71)	0	(0.58)
156	3	17.9846		1	
157	1	7.2622		1	
158	1	6.7354	7.25	1	1.00
159	1	7.8894	(0.48)	1	(0.00)
160	1	7.1311		1	

NB: 0: No Attack / 1: Attempted Attack / 2: Slight Attack / 3: Average Attack / 4: Strong Attack

Table XIII

Specimen No.	Solution Conc. (%) Eq.	Termite Attack Rating
163	10	1
165	7	4
172	5	4
174	3	4
179	1	4

NB: 0: No Attack / 1: Attempted Attack / 2: Slight Attack / 3: Average Attack / 4: Strong Attack

Table VIIIa

Specimen No.	Tunneling Activity	Visible Mortality
189	Yes	1-33%
190	Yes	1-33%
191	Yes	1-33%
192	Yes	1-33%

Table IXa

Specimen No.	Tunneling Activity	Visible Mortality
185	Yes	1-33%
186	Yes	1-33%
187	Yes	1-33%
188	Yes	1-33%

Table Xa

Specimen No.	Tunneling Activity	Visible Mortality
41	Yes	1-33%
42	Yes	1-33%
43	Yes	1-33%
44	Yes	1-33%
45	Yes	1-33%
46	Yes	1-33%
47	Yes	1-33%
48	Yes	1-33%
49	Yes	1-33%
50	Yes	1-33%
51	Yes	1-33%
52	Yes	1-33%
53	Yes	1-33%
54	Yes	1-33%
55	Yes	1-33%
56	Yes	1-33%
57	Yes	No
58	Yes	No
59	Yes	No
60	Yes	No

Table XIa

Specimen No.	Tunneling Activity	Visible Mortality
121	Yes	67-100%
122	Yes	67-100%
123	Yes	67-100%
124	Yes	67-100%
125	Yes	67-100%
126	Yes	67-100%
127	Yes	67-100%
128	Yes	67-100%
129	Yes	67-100%
130	Yes	67-100%
131	Yes	67-100%
132	Yes	67-100%
133	Yes	34-66%
134	Yes	34-66%
135	Yes	34-66%
136	Yes	34-66%
137	Yes	34-66%
138	Yes	34-66%
139	Yes	34-66%
140	Yes	34-66%

5

Table XIIa

Specimen No.	Tunneling Activity	Visible Mortality
141	Yes	34-66%
142	Yes	67-100%
143	Yes	67-100%
144	Yes	34-66%
145	Yes	67-100%
146	Yes	67-100%
147	Yes	100%
148	Yes	67-100%
149	Yes	67-100%
150	Yes	67-100%
151	Yes	67-100%
152	Yes	67-100%
153	Yes	34-66%
154	Yes	34-66%
155	Yes	67-100%
156	Yes	67-100%
157	Yes	67-100%
158	Yes	67-100%
159	Yes	67-100%
160	Yes	67-100%

Table XIIIa

Specimen No.	Tunneling Activity	Visible Mortality
163	Yes	1-33%
165	Yes	1-33%
172	Yes	1-33%
174	Yes	1-33%
179	Yes	1-33%

From the results reported above the following conclusions can be reached. The test results for TBBA, TBBF and TBBS indicate a very

significant effect in restricting termite damage to timber specimens treated with these actives.

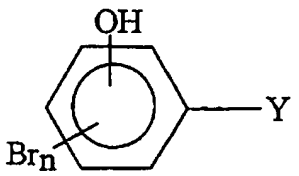
- TBBA, TBBF and TBBS all display significant termiticidal and termite repellent activity. Timber impregnated with these actives is protected against termite attack.

All the above description of preferred embodiments has been given for the purpose of illustration and is not meant to limit the invention. Many variations and modifications can be provided by the skilled person, all which are meant to be encompassed by the claims to follow.

Claims

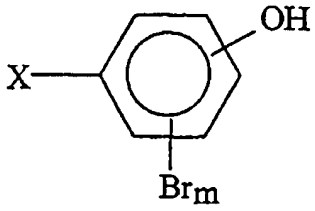
1. An insecticidal and termiticidal wood preservative composition containing as an active ingredient a compound of the formula:

(I)



wherein Y is a moiety selected from halogen, substituted aromatic ring and/or aliphatic residue, and n= 1 to 4.

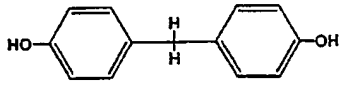
2. A composition according to claim 1, wherein Y is



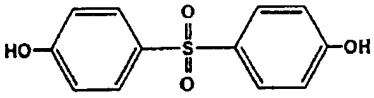
wherein X is a moiety selected from alkyl, S or SO_2, and m= 0 to 4.

3. A composition according to claim 1 or 2, wherein the compound of Formula (I) is selected from among tetrabromobisphenol A (TBBA)

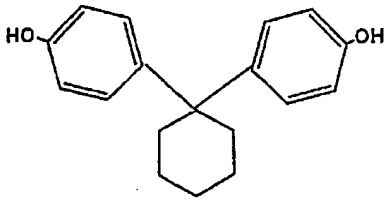
[4,4'-isopropylidenebis(2,6-dibromophenol)], tribromobisphenol A [2,2',6-tribromo-4,4'-isopropylidenediphenol] (TBA) or a bromine derivative of a compound selected from:



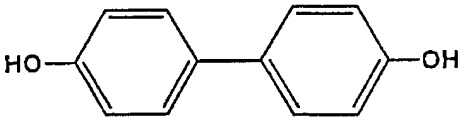
Bisphenol F, Bis(4-hydroxyphenyl)methane



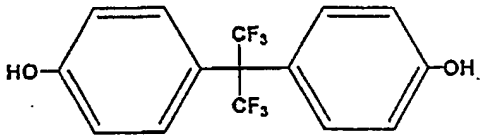
Bisphenol S, 4,4'-sulfonyldiphenol



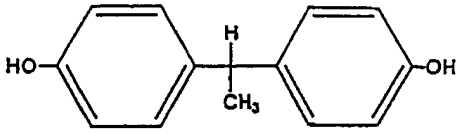
Bisphenol Z, 4,4'-cyclohexylidenediphenol



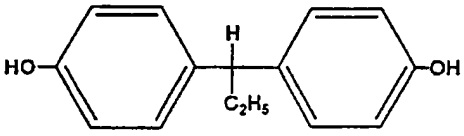
4,4'-Biphenol



2,2-Hexafluoroisopropylidinediphenol

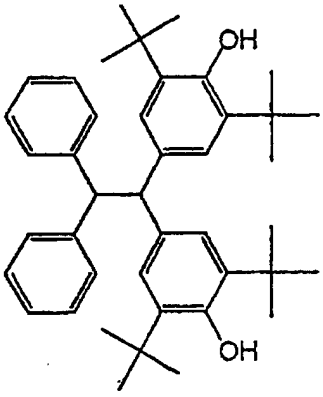


1,1-Ethylidenediphenol



1,1-propylidenediphenol

1,1,2,2-tetra-(4-hydroxyphenyl)ethane, or



4. A composition according to claim 1, which is dissolved in a solvent.

5. A composition according to claim 4, wherein the solvent is ethanol.

6. A composition according to claim 4, wherein the solvent is an aqueous basic solution.

7. A composition according to claim 6, wherein the base is NaOH.

8. A composition according to any one of claims 1 to 7, wherein the active material is TBBA and its concentration ranges between 1% (w/w) and 10% (w/w).

9. A composition according to any one of claims 1 to 8, further comprising conventional termiticidal and insecticidal agents.

10. A method for protecting wood against termites, comprising impregnating the wood with a wood preservative composition according to any one of claims 1 to 9.

11. Use of a composition according to any one of claims 1 to 9, for protecting wood against termites.

12. A composition according to any one of claims 1 to 9, for use as a termiticidal agent, alone or together with conventional termiticidal and/or insecticidal agents.

INTERNATIONAL SEARCH REPORT

Int ial Application No
PCT/IL 02/00545

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B27K3/40 A01N31/16 A01N31/08 A01N41/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B27K A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
CHEM ABS Data, EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2 455 652 A (BRALLEY JAMES A ET AL) 7 December 1948 (1948-12-07) example 5	3-9
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; KANTAM, MANNEPALLI LAKSHMI ET AL: "Bromination process and catalysts for the preparation of tetrabromobisphenol A" retrieved from STN Database accession no. 135:20085 XP002215005 abstract & US 6 245 950 B1 (KANTAM, MANNEPALLI LAKSHMI ET AL) 12 June 2001 (2001-06-12)	3-9

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 September 2002	12/11/2002

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Marie, G

Form PCT/ISA/210 (second sheet) (July 1992)

BNSDOCID: <WO_ 03009981A1_I_>

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; MANIMARAN, THANIKAVELU ET AL: "Manufacture of tetrabromobisphenol A" retrieved from STN Database accession no. 133:351788 XP002215006 abstract & US 6 147 264 A (MANIMARAN, THANIKAVELU ET AL) 14 November 2000 (2000-11-14)	3-9
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; HIGA, TATSUO ET AL: "Thelepin, a new metabolite from the marine annelid Thelepus setosus" retrieved from STN Database accession no. 81:75098 XP002215007 abstract & J. AMER. CHEM. SOC. (1974), 96(7), 2246-8 ,	3-9
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; RAJKOTIA, K. M. ET AL: "Synthesis and biological activities of 1,1'-bis(p-substituted-benzoyloxy-substituted-phenyl)cyclohexane" retrieved from STN Database accession no. 130:3662 XP002215008 abstract & JOURNAL OF THE INDIAN CHEMICAL SOCIETY (1998), 75(9), 524-525 ,	3-9
A	EP 1 018 413 A (BAYER AG ;KEMIHOLZ CO LTD (JP); OSHIKA SHINKO CO (JP)) 12 July 2000 (2000-07-12) the whole document	1-12

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

Claims Nos.: 1-2

The intended use is not regarded as a technical feature in composition claims, therefore present claims 1-9 and 12 are regarded are compositions per se.
Claims 1,2 and 12 relate to an extremely large number of possible compositions. Support within the meaning of Article 6 PCT and disclosure within the meaning of Article 5 PCT is to be found, however, for only a very small proportion of said compositions claimed. In the present case, the claims so lack support, and the application so lacks disclosure, that a meaningful search over the whole of the claimed scope is impossible. Consequently, the search has been carried out for those parts of the claims which appear to be supported and disclosed, namely those parts relating to the compositions per se as defined in claim 3.
The subject-matter of claims 10 and 11, which are directed to an activity, has not been searched for the compositions as defined in claim 1. Indeed, a complete search was not possible in that case for overflow reasons.

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

INTERNATIONAL SEARCH REPORT

national application No.
PCT/IL 02/00545

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☒ Claims Nos.: 1-2
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
see FURTHER INFORMATION sheet PCT/ISA/210

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

BNSDOCID: <WO_03009981A1_I_>>

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